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NEUTRON AND SYNCHROTRON X-RAY SCATTERING STUDIES OF SUPERCONDUCTORS

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ABSTRACT

Superconductors hold the promise for a more stable and efficient electrical grid, but new isotropic, high-temperature superconductors are needed in order to reduce cable manufacturing costs. The effort to understand high-temperature superconductivity, especially in the layered cuprates, provides guidance to the search for new superconductors. Neutron scattering has long provided an important probe of the collective excitations that are involved in the pairing mechanism. For the cuprates, neutron and x-ray diffraction techniques also provide information on competing types of order, such as charge and spin stripes, that appear to be closely connected to the superconductivity. Recently, inelastic x-ray scattering has become competitive for studying phonons and may soon provide valuable information on electronic excitations. Examples of how these techniques contribute to our understanding of superconductivity are presented.

1. INTRODUCTION

With increasing use of renewable energy sources, the electric grid will continue to be crucial for moving electric power from generating sites to customers. In the U.S., improving the stability of the grid is as important as increasing capacity; a dynamic imbalance between reactive and resistive loads can lead to events such as the blackout that affected much of the northeastern U.S. on 14 August 2003. A good discussion of the application of high-temperature superconductors to power cables and to devices such as the dynamic synchronous condenser is given by Malozemoff, Mannhart, and Scalapino (2005). Some of the important research questions relevant to developing improved and practical superconductors are described in a workshop report by Basic Energy Sciences, U.S. Dept. of Energy (2006). Scattering studies, in particular, can contribute to understanding the mechanism of high-temperature superconductivity, which, in turn, can provide direction to the search for new superconducting materials with enhanced properties.

Neutron scattering has long provided crucial information about the collective excitations

involved in the electronic pairing mechanism required for superconductivity. For example, the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity (Bardeen, Cooper, and Schrieffer, 1957) is based on the idea that electrons pair by exchanging phonons. The first direct identification of the spectrum of excitations responsible for pairing, $\alpha^2 F(\omega)$, was made by McMillan and Rowell (1965) for a Pb-insulator-Pb tunnel junction; they extracted the spectrum by analyzing the second derivative of the tunnel current with respect to bias voltage. They were able to easily identify the spectrum as the density of states for Pb phonons because Brockhouse, Arase, Caglioti, Rao, and Woods (1962) had previously done a beautiful job of measuring the phonon dispersions for a Pb crystal by neutron scattering with a triple-axis spectrometer (TAS).

It is also possible to see the impact of superconductivity by measuring phonon line widths. In the normal state, a phonon can decay by exciting electron-hole pairs; however, in the superconducting state, a phonon with an energy less than twice the superconducting energy gap, Δ , cannot decay in the normal fashion because it does not have enough energy to break up an electron pair. Thus, the line width for phonons with $\hbar\omega < 2\Delta$ should decrease on cooling into the superconducting state (and increase for $\hbar\omega \gtrsim 2\Delta$ due to an enhanced density of electronic states). Shapiro, Shirane, and Axe (1975) demonstrated this nicely for a crystal of Nb using a TAS. Quite recently, there have been new experiments in this direction using the enhanced energy resolution of a TAS with resonant spin echo. Measurements on Pb and Nb reveal new features that go beyond the conventional BCS theory (Aynajian, Keller, Boeri, Shapiro, Habicht, and Keimer, 2008).

2. CUPRATES

High-temperature superconductivity in the cuprates is achieved by doping holes into antiferromagnetic CuO_2 planes. The electronic doping of carriers converts the insulating parent compounds into unusual metallic phases. Various studies indicate that the “normal” metallic state does not contain coherent quasiparticles, the latter being a prerequisite for the application of the conventional BCS theory. Many theorists believe that antiferromagnetic interactions may be important for pairing of carriers in the superconducting state, although there is no consensus on how this occurs. Scattering studies have been quite important in determining how the antiferromagnetic correlations evolve with doping and temperature. [For more detailed reviews of the neutron scattering work, please see Birgeneau, Stock, Tranquada, and Yamada (2006) and Tranquada (2007).]

2.1 Antiferromagnetic order and spin waves.

Antiferromagnetic order in the parent compounds La_2CuO_4 and $\text{YBa}_2\text{Cu}_3\text{O}_6$ was discovered by neutron diffraction on polycrystalline samples (Vaknin, Sinha, Moncton, Johnston, Newsam, Safinya, and H. E. King, 1987; Tranquada, Cox, Kunnmann, Moudén, Shirane, Suenaga, Zolliker, Vaknin, Sinha, Alvarez, Jacobsen, and Johnston, 1988). This work demonstrated that the Cu atoms in the CuO_2 planes have magnetic moments, corresponding approximately to spin $\frac{1}{2}$, that order in a simple collinear antiferromagnetic structure. Typical Néel temperatures, T_N , of insulating cuprates are in the range of 250–500 K; however, these temperatures do not give a meaningful measure of the nearest-neighbor superexchange energy, J , as the ordering is limited by the very weak coupling between planes.

When single crystals of La_2CuO_4 became available, measurements of energy-integrated magnetic scattering demonstrated that the Cu moments maintain antiferromagnetic correlations up to $T \gg T_N$ (Shirane, Endoh, Birgeneau, Kastner, Hidaka, Oda, Suzuki, and Murakami,

1987), reflecting a large J . A more direct measure of J is obtained by measuring the spin waves. Measurements using TASs confirmed the large spin-wave velocity; however, the full bandwidth of $2J$ was too large to be fully characterized with thermal neutrons. A complete measurement of the spin-wave spectrum had to await the development of time-of-flight chopper instruments at spallation sources, such as the MAPS spectrometer at ISIS, Rutherford-Appleton Lab, in the UK. [Similar instruments will soon become available at the Spallation Neutron Source (SNS) in the US and at the Japanese Proton Accelerator Research Complex (J-PARC).] For example, a thorough study of the spin waves in La_2CuO_4 was performed by Coldea, Hayden, Aeppli, Perring, Frost, Mason, Cheong, and Fisk (2001). Analysis of the dispersion at high energies along the zone boundary demonstrated the need to include 4-spin cyclic exchange in the effective spin Hamiltonian.

2.2 Stripe order.

To obtain superconductivity, one must dope holes into the the antiferromagnetic CuO_2 planes. Inelastic neutron scattering studies demonstrated that the antiferromagnetic correlations survived into the superconducting regime (Bourges, Regnault, Henry, Vettier, Sidis, and Burlet, 1995; Kastner, Birgeneau, Shirane, and Endoh, 1998). This observation created a puzzle, as the antiferromagnetic correlations appear to derive from the correlated insulator state while the doped carriers, at sufficient density, are mobile. How could these two contradictory behaviors coexist? One solution came with the discovery of charge and spin stripe order.

Stripe order was discovered through neutron diffraction measurements on a single crystal of $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$ (Tranquada, Sternlieb, Axe, Nakamura, and Uchida, 1995), and has been confirmed more recently in $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ (Fujita, Goka, Yamada, Tranquada, and Regnault, 2004). These compounds have almost the same crystal structure as superconducting $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, except for a subtle difference associated with the tilt direction of the CuO_6 octahedra (Axe, Moudden, Hohlwein, Cox, Mohanty, Moodenbaugh, and Xu, 1989). This small difference causes Cu-O bonds in orthogonal directions within a CuO_2 plane to be inequivalent. It is this rotational anisotropy that can pin the charge modulation which is revealed by the presence of very weak superlattice peaks split about the fundamental Bragg peaks. In addition, there are magnetic superlattice peaks split about the antiferromagnetic wave vector; these are essentially in the same positions as the low-energy magnetic excitation peaks first observed in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ by Cheong, Aeppli, Mason, Mook, Hayden, Canfield, Fisk, Clausen, and Martinez (1991). The orientations and magnitudes of the ordering wave vectors indicate that the charge order has a period of four lattice spacings along a Cu-O bond direction, while the magnetic period is twice that. In essence, there are narrow antiferromagnetic domains between the charge stripes, with the orientation of the spins flipping direction on crossing a charge stripe. The nature of the stripe order is consistent with some theoretical predictions for doped antiferromagnets (Zaanen and Gunnarsson, 1989; Machida, 1989; Kivelson, Bindloss, Fradkin, Oganessian, Tranquada, Kapitulnik, and Howald, 2003).

The nature of stripe order has been somewhat controversial, and a number of other techniques have helped in establishing its character. For example, measurements with polarized neutron have demonstrated that, to the extent the modulation is unidirectional, the ordered spins have a collinear arrangement (Christensen, Rønnow, Mesot, Ewings, Momono, Oda, Ido, Enderle, McMorro, and Boothroyd, 2007); thus, the magnetic superlattice peaks cannot be explained by a pure spiral structure. Synchrotron techniques have also played a valuable role. Diffraction with a high-energy (100 keV) x-ray beam at HASYLAB has proven to be an effective way to characterize the charge order peaks. Such measurements demonstrated that the charge-stripe structure is modulated along the c -axis (perpendicular

to the planes), consistent with a rotation of the stripe direction by 90° from one layer to the next (driven by the underlying crystal symmetry), together with Coulomb repulsion between stripes in equivalent, next-nearest-neighbor planes (v. Zimmermann, Vigliante, Niemöller, Ichikawa, Frello, Uchida, Andersen, Madsen, Wochner, Tranquada, Gibbs, and Schneider, 1998).

The neutron and x-ray diffraction measurements of charge-order-related superlattice peaks essentially provide indirect information about the charge order via the induced lattice modulation. A direct measure of the charge order has recently become available with the development of soft-x-ray resonant diffraction. This technique has been applied both to $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ (Abbamonte, Rusydi, Smadici, Gu, Sawatzky, and Feng, 2005) at the NSLS and to $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$ (Fink, Schierle, Weschke, Geck, Hawthorn, Wadati, Hu, Durr, Wizent, Büchner, and Sawatzky, 2008) at BESSY. Here one gains sensitivity to partially filled states at the Fermi level by scanning the photon energy through the O K edge (at 530 eV) and the Cu L_3 edge (at 930 eV). While one can quibble over whether there is a true modulation of *charge* density, these measurements clearly demonstrate a spatial modulation of the occupancy of O $2p_\sigma$ orbitals. It is worth noting that one can also detect magnetic order with soft x-ray resonant diffraction at L edges, as has been demonstrated for stripe order in $\text{La}_{1.8}\text{Sr}_{0.2}\text{NiO}_4$ (Schussler-Langeheine, Schlappa, Tanaka, Hu, Chang, Schierle, Benomar, Ott, Weschke, Kaindl, Friedt, Sawatzky, Lin, Chen, Braden, and Tjeng, 2005).

2.3 Spin dynamics.

As already mentioned, the MAPS time-of-flight spectrometer has proven to be quite powerful for the measurement of spin-wave spectra in two-dimensional antiferromagnets. It has also been useful for measuring the effective dispersions of spin fluctuations in superconducting samples. Examples include results for underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (Hayden, Mook, Dai, Perring, and Doğan, 2004; Stock, Buyers, Cowley, Clegg, Coldea, Frost, Liang, Peets, Bonn, Hardy, and Birgeneau, 2005; Hinkov, Bourges, Pailhes, Sidis, Ivanov, Frost, Perring, Lin, Chen, and Keimer, 2007), optimally-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (Vignolle, Hayden, McMorro, Rønnow, Lake, Frost, and Perring, 2007), and stripe-ordered $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ (Tranquada, Woo, Perring, Goka, Gu, Xu, Fujita, and Yamada, 2004). Comparison of these and other results suggest that there is a universal dispersion for the cuprates, with an hour-glass shape. The waist of the hour glass has an energy in the range of 30–50 meV for samples ranging from mildly-underdoped to optimally doped. For underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, it is found that this energy scale (also called E_{cross}) varies linearly with x for $x \lesssim \frac{1}{8}$ (Matsuda, Fujita, Wakimoto, Fernandez-Baca, Tranquada, and Yamada, 2008).

For stripe-ordered $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$, the magnetic spectrum can be understood as fluctuations about the magnetically-ordered ground state. The essential features of this spectrum change little when the stripe *order* is eliminated by warming (Xu, Tranquada, Perring, Gu, Fujita, and Yamada, 2007), consistent with the concept that dynamic stripe correlations remain even in the absence of order. The fact that a very similar magnetic spectrum is observed in superconducting samples suggests that dynamic stripes may be a common feature of the superconducting cuprates. Also consistent with this idea is the fact that the incommensurability of the low-energy spin fluctuations in underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ varies with doping in the manner one would expect if adding holes simply increases the density of charge stripes (Yamada, Lee, Kurahashi, Wada, Wakimoto, Ueki, Kimura, Endoh, Hosoya, Shirane, Birgeneau, Greven, Kastner, and Kim, 1998).

Of course, the charge-stripe density cannot increase forever, and experimentally the effective density, as indicated by the magnetic incommensurability in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, saturates

for $x \gtrsim \frac{1}{8}$. Since it is not energetically favorable for the hole density within the stripes to increase, the extra holes must go into uniformly-doped regions, coexisting with stripe-correlated patches. As discussed by Birgeneau et al. (2006), there is evidence for phase separation in overdoped cuprates. If antiferromagnetic correlations, characteristic of the parent insulator, can only exist in the stripe-correlated regions, and the areal density of these regions decreases with overdoping, then we would also expect the spectral weight of antiferromagnetic fluctuations to decline. Indeed, this is just what has been observed in recent inelastic neutron scattering studies (Wakimoto, Yamada, Tranquada, Frost, Birgeneau, and Zhang, 2007; Lipscombe, Hayden, Vignolle, McMorro, and Perring, 2007).

To test the generality of results obtained so far, studies are being pursued on cuprates such as $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Fauque, Sidis, Capogna, Ivanov, Hradil, Ulrich, Rykov, Keimer, and Bourges, 2007) where sample size can pose a challenge to accurately extracting the magnetic signal. Recent success in growing large crystals of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Wen, Xu, Xu, Hücker, Tranquada, and Gu, 2008) has led to enhanced signal; new results will be published in the near future.

Before leaving this section, it is worth noting that polarized neutrons can be quite useful for distinguishing spin fluctuations from phonons. There is not a lot of published work in which this technique has been applied to layered cuprates; however, a good example is given by Regnault, Rønnow, Lorenzo, Bellissent, and Tasset (2003), who separated the magnetic cross section from the phonon background in $\text{YBa}_2\text{Cu}_3\text{O}_{6.85}$. The reason for not using this approach more routinely is that one must pay a significant cost in intensity in order to work with polarized neutrons.

2.4 Phonons.

Phonons may play an indirect role in cuprate superconductivity, as they provide a channel for screening charge modulations and fluctuations. Inelastic neutron scattering is the traditional technique for measuring phonon dispersions. In each cuprate family, there are many phonon branches because of the large number of atoms in the typical unit cell. Each branch has a different \mathbf{Q} -dependent structure factor, so one generally needs to employ model calculations to help in making proper identifications.

It took considerable accumulated experience with the cuprates to understand what features are normal and which are anomalous. Eventually it has been learned that most branches are well described by conventional model calculations, such as ones based on the local-density approximation for the electronic structure (Pintschovius, 2005).

The branch one might expect to interact most strongly with the conduction electrons is the longitudinal bond-stretching mode. The higher-energy mode of this type involves mostly oxygen motion, oscillating between neighboring Cu atoms. This mode is fairly flat in the insulating parent compounds; however, the zone-boundary energy softens with doping as a consequence of electronic screening (Stercel, Egami, Mook, Yethiraj, Chung, Arai, Frost, and Dogan, 2008).

The interesting behavior occurs approximately halfway to the zone boundary, where a rapid drop in energy and a large broadening are observed in superconducting samples (Reznik, Pintschovius, Ito, Iikubo, Sato, Goka, Fujita, Yamada, Gu, and Tranquada, 2006). This anomaly is also quite strong in stripe-ordered $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ and $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$; however, it disappears with extreme under- or over-doping.

While inelastic neutron scattering with a TAS remains an excellent way to measure phonons, inelastic x-ray scattering has become a competitive technique (Uchiyama, Baron, Tsutsui, Tanaka, Hu, Yamamoto, Tajima, and Endoh, 2004; Fukuda, Mizuki, Ikeuchi, Yamada, Baron, and Tsutsui, 2005). While x-rays do not offer in any particular time efficiency, they do make it possible to study much smaller crystals.

2.5 Relevance of stripes to superconductivity.

While certain signatures consistent with stripe correlations seem to be common to cuprate superconductors, there has been a wide spread belief that stripe order is inherently bad for superconductivity. This belief is reinforced by the fact that the bulk T_c is very low in samples with the highest stripe-ordering temperatures (Tranquada, Axe, Ichikawa, Moodenbaugh, Nakamura, and Uchida, 1997). To test the relevance of stripes to superconductivity requires turning to some experimental measurements complementary to the scattering techniques that we have focused on so far.

Photoemission and tunneling measurements on stripe-ordered $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ provide evidence for a d -wave-like gap, similar to that found in superconducting cuprates (Valla, Federov, Lee, Davis, and Gu, 2006). Optical conductivity provides further evidence for a gap that develops with the onset of charge-stripe order (Homes, Dordevic, Gu, Li, Valla, and Tranquada, 2006). Those results motivated a careful study of transport properties and magnetic susceptibility (Li, Hucker, Gu, Tsvelik, and Tranquada, 2007). It was discovered that the in-plane resistivity drops by an order of magnitude at the spin-stripe-ordering temperature (40 K), whereas the c -axis resistivity shows no evidence of this transition and grows on cooling through it. Very weak diamagnetism sets in at the same temperature, but only for a magnetic field applied perpendicular to the planes (resulting in screening currents within the planes). The in-plane resistivity eventually goes to zero at 16 K (while the c -axis resistivity remains finite), well above the bulk diamagnetic transition at 5 K. These results (plus a few more) have been interpreted as evidence for the onset of 2D superconducting correlations at 40 K (a temperature higher than the bulk T_c of any composition in the $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ phase diagram), with long-range 2D superconducting order setting in at 16 K (through a Berezinskii-Kosterlitz-Thouless transition).

The conclusion regarding 2D superconductivity coexisting with stripe order supports the idea that stripes are good for pairing and for superconductivity within the CuO_2 planes; however, it creates a new puzzle. All of the cuprate superconductors are effectively 2D superconductors, but Josephson coupling between the planes inevitably leads to 3D superconductivity. In order to get 2D behavior, one needs a mechanism to frustrate the interlayer Josephson coupling. Two groups have independently proposed a model based on the idea of a sinusoidally-modulated superconducting wave function that has a large magnitude at the positions of the charge stripes and goes to zero in the middle of the antiferromagnetic domains (Berg, Fradkin, Kim, Kivelson, Oganessian, Tranquada, and Zhang, 2007; Himeda, Kato, and Ogata, 2002). As mentioned earlier, scattering studies indicate that the stripe direction rotates 90° from one layer to the next; this should cause the orientation of the modulated superconducting state to rotate as well. The Josephson coupling between these orthogonal modulations will average to zero, thus leading to the 2D response.

While the unusual superconducting state proposed for $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ is different from the uniform d -wave state believed to occur in most cuprate superconductors, it nevertheless indicates an intimate connection between stripe correlations and pairing. While there is much more work required to achieve a consensus understanding of the superconductivity in cuprates, it appears that the information provided by scattering techniques will be essential

to the final story.

3. Future

There is a new flood of excitement now with the recent discovery of high-temperature superconductivity in $\text{LaFeAsO}_{1-x}\text{F}_x$ ($T_c = 43$ K) by Takahashi, Igawa, Aii, Kamihara, Hirano, and Hosono (2008). An onset temperature for superconductivity as high as 55 K has been reported in the case where La is replaced by Sm (Ren, Lu, Yang, Yi, Shen, Zheng-Cai, Che, Dong, Sun, Zhou, and Zhao, 2008). These compounds contain FeAs layers, with tetrahedrally-coordinated Fe sites, alternating with RO layers, where R is La or a rare earth.

This system is especially exciting because it seems to be more flexible for substitutions than the cuprates. For example, low-temperature superconductivity was first observed in a variant with P in place of As (Kamihara, Hiramatsu, Hirano, Kawamura, Yanagi, Kamiya, and Hosono, 2006), and now it has been shown that superconductivity can also be achieved with As replaced by Bi and Fe replaced by Ni (Kozhevnikov, Leonidova, Ivanovskii, Shein, Goshchitskii, and Karkin, 2008). Furthermore, one can even get rid of the oxygen, as it has now been found that $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ is superconducting with $T_c = 38$ K (Rotter, Tegel, and Johrendt, 2008). This variability offers considerable promise for the discovery of new superconductors in the future.

Neutron scattering is already being applied to study structural and magnetic transitions in these materials. For example, it has been found that nonsuperconducting LaFeAsO lowers its symmetry from tetragonal to monoclinic on cooling through 155 K, and then undergoes an antiferromagnetic ordering at 137 K, achieving a maximum ordered moment of just $0.4 \mu_B$ per Fe atom (de la Cruz, Huang, Lynn, Li, Ratcliff, Zarestky, Mook, Chen, Luo, Wang, and Dai, 2008). Doping with enough F to raise T_c to 26 K appears to eliminate both the structural transition and the antiferromagnetic ordering. Interestingly, switching from La to Nd, one finds that NdFeAsO has a similar structural transition, but antiferromagnetic ordering, involving both Nd and Fe moments, occurs only below 2 K (Qiu, Bao, Huang, Lynn, Yildirim, Simmons, Gasparovic, Li, Green, Wu, Wu, and Chen, 2008).

There is a great deal of theoretical interest in these materials, with many predictions that magnetic correlations play a role in the superconductivity. As single crystals become available, neutron scattering will inevitably be used to study dynamic spin correlations and their temperature dependence through the superconducting transition.

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